Characteristics of Tropospheric Ozone Profile and Implications on the Origin of Ozone over Subtropical China in Spring 2001

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Abstract

During the TRACE-P (TRAnsport and Chemical Evolution over the Pacific) period in spring 2001, we launched ozonesonde at three subtropical Chinese sites at Kunming (102.68^{o}E, 25.03^{o}N), Hong Kong (114.17^{o}E, 22.31^{o}N) and LinAn (119.75^{o}E, 30.30^{o}N). The sites extend from the southwestern China close to the Southeast (SE) Asia border to southeastern Asia coast and the edge of subtropics of central-eastern China respectively. The aims are to provide ozonesonde data within the source regions of Chinese mainland and to investigate the source of tropospheric ozone (O_3) and to what extent the SE Asian biomass burning emissions impacting on the tropospheric O_3 over subtropical Chinese mainland and O_3 outflows to the Pacific. The results showed that there are substantial variations in the vertical O_3 distributions over these subtropical sites with low O_3 values at the upper troposphere of Hong Kong, high O_3 values at the middle and upper troposphere of LinAn and frequent O_3 enhancements at the lower troposphere of Hong Kong and Kunming. The low values over the upper troposphere of HK in spring 2001 was not usually observed from 1993 to 2000 and is as a result of the transport of O_3-depleted air from the inter-tropical convergence zone of equatorial SE Asian regions following the East Asia local Hadley circulation. Such transport process does not affect the higher latitude at the edge of subtropics of LinAn where stratospheric O_3 is the major contributing source to the middle and upper tropospheric $O_{3.}$ The O_3 enhancements over the lower troposphere of Kunming and Hong Kong are caused by the SE Asian biomass burning emissions. Such enhancements are frequently observed over Hong Kong, less often over Kunming and scarcely over LinAn. Our analysis showed the biomass burning emissions from SE Asia in spring 2001 mainly affected the southern parts of subtropical Chinese region.

INTRODUCTION

One of the objectives of TRACE-P is to better understand the chemical evolution of tropospheric O_3 associated with Asian outflow over the western Pacific and the processes that control this evolution [*Jacob et al.*, this issue]. During March-April 2001, when the two NASA aircrafts were flying over the western Pacific, we had performed simultaneous ozonesonde launching at three subtropical locations in South China. The first aim is to supplement the aircraft measurements by providing ozonesonde data within the source regions of the Asian Continent inside mainland China. The second aim is to investigate the source of tropospheric O_3 and to find the extent of the impact of SE Asian biomass burning emissions on tropospheric O_3 over subtropical China and on O_3 outflow to the Pacific.

The large-scale air pollutant emissions in the Indonesian region of SE Asia and South Asia as a result of forest fire, biomass and biofuel burning emissions has drawn international focus recently. This pollution is strongly impacting on the chemical composition of the atmosphere and on the climate system [*Fujiwara et al.*, 1999, *Matsueda and Inoue*, 1999, *Tsutsumi et al.*, 1999, *Chan at al.*, 2001, *Lelieveld et al.*, 2001, *Thompson et al.*, 2001, *UNEP and* C^4, 2002]. The possible impact of biomass burning emissions from the SE Asia subcontinent on tropospheric O_3 over subtropical Chinese mainland was first suggested by *Liu et al.* [1999], who noted a regularly substantial enhancement of tropospheric O_3 in the springtime over the lower troposphere (2.0 - 4.5 km) of Hong Kong. *Chan et al.* [2000] showed with a case study that the O_3 enhancements are the result of photochemical O_3 produced by biomass burning emissions from the Indo-Burma region of SE Asian subcontinent. *Chan et al.* [2002] revealed through satellite data analysis that the biomass burning activities in the

the Indo-Burma and Indian-Nepal regions reach their full strengths in the dry season from late February to middle April coinciding with the springtime tropospheric O_3 maximum over Hong Kong [*Chan et al.*, 1998]. The chemical speciation and transport characteristics analysis of the O_3-rich air masses reaching Hong Kong showed that these air masses traverse over a large geographical region of subtropical Chinese mainland before outflowing to the Pacific [*Chan et al.*, 2002]. These findings inspire us to explore the possible impacts of the biomass burning emissions from SE Asia on the tropospheric O_3 over the general subtropical Chinese regions and on the outflows to the Pacific.

Talbot et al. [1996] found that the strong cyclonic activities over Asia during the 1994 Pacific Exploratory Mission (PEM)-West A experiment (September – October 1991) could have transported the biomass burning emissions from SE Asia or emissions from the extensive use of biomass materials to the upper troposphere. *Blake et al.* [1997] noted that the polluted air masses observed over the western Pacific during the 1994 PEM-West B experiment are rich in biomass burning emission tracers, including methyl chloride and other hydrocarbon (HC) and halogen HC tracers. *Folkins et al.* [1997] showed that deep convection can inject emissions from SE Asian biomass burning to near tropical tropopause. *Harris et al.* [1998] pointed out with evidence from the CO, O_3 and CH_4 relationship that the strong horizontal O_3 gradient from the SE Asian continent to the Mauna Loa Observatory (MLO) at central Pacific is related to the springtime tropospheric O_3 maximum observed at the MLO station. However the authors cannot identify the sources contributing to the O_3 gradient. *Chan et al.* [2002] showed with forward trajectory that the O_3-rich air masses observed over Hong Kong and outflowed to the western and central Pacific from the subtropical Asian coast can reach the western coast of the United States within ten days. In this paper, we present the ozonesonde measurement data observed at three South China stations. The objective is to investigate the characteristics of tropospheric O_3 profiles and explore their origin over the large geographical regions of subtropical Chinese mainland.

EXPERIMENT

During the TRACE-P period when the two NASA aircrafts were flying over the East Asian coast and the western Pacific, ozonesonde was launched at three subtropical Chinese sites at Kunming, Hong Kong and LinAn. These sites cover the region from southwestern China close to the SE Asian border to the southeastern Asian coast and to the edge of the subtropics at central-eastern China (Figure 1). The LinAn launching site (119.75^{o}E, 30.30^{o}N) is primarily a rural area. It is 134 meters above sea level (ASL). There were a total of 28 launches from March 3 to April 13, 2001 with a trial launch in February 21. The O_3 profiles were available everyday from March 3 to 13. After that there was at least a profile every other day until April 13 with an exception from April 4 to 7, during which only two profiles were available. The Kunming launching site (102.68^{o}E, 25.03^{o}N) is situated on the edge of the Tibet Plateau at a suburban area with an altitude of 1820 meters ASL. There were a total of 27 launches from March 1 to April 13 with a trial launch on February 28, 2001. The launching site in Hong Kong is the Upper Air Monitoring Station of the Hong Kong Observatory. A detailed description on the launching site in Hong Kong (114.17^{o}E, 22.31^{o}N) can be found in *Chan et al.* [1998]. The launching in Hong Kong for the TARCE-P mission started from March 2000 until June 2001 with a frequency of once per week and higher frequencies when the two aircrafts flew close to the Hong Kong region. In all three sites, the launching was performed at 5:00 to 6:00 UT (13:00 to 14:00 LST).

Tropospheric ozone profiles were measured by in-situ electrochemical concentration cell (ECC) ozonesondes (Model 6a, Vaisala) that were coupled with a standard radiosonde (RS80-15GE, Vaisala). A Vaisala DigiCora MW 15 receiving system was used. The ozonesonde launching system used, experimental procedures, quality control and assurance procedures were similar to those described by *Chan et al.* [1998] and *Chan et al.* [2002]. The ECC ozonesonde has been used since the early 1970s. The ozone data measured by ozonesonde have precisions from ± 3 to ± 12% in the troposphere, ± 3% in the stratosphere up to 10 mbar altitude and ± 10% at 4 mbar altitude. The corresponding accuracies for individual ozonesonde sounding are ± 6% near the ground, 7 to 17% in the high troposphere, where ozone mixing ratios are low, 5% in the low stratosphere up to about 10 mbar altitude and 6 to 14% at 4 mbar altitude [*Komhyr et al.*, 1995]. The integrated total column ozone from the ozonesonde compares well with that measured by Dobson Spectrophotometer with an average ratio of 1.04 ± 0.05 between the former and the latter [*Oltmans et al.*, 2001].

RESULT AND DISCUSSION

Characteristic of Tropospheric Ozone Profiles

Figure 2 shows the average profiles of O_3 mixing ratio, water mixing ratio, potential temperature, wind speed and wind direction below 18 km in the three launching sites. The average profiles comprise all the profiles available during the TRACE-P period. Comparing the three O_3 profiles, we noted there are substantial differences in the distributions of O_3 in subtropical atmospheres of South China. At the edge of the subtropics over LinAn, O_3 mixing ratio showed a steady increase from less than 50 ppbv close to the surface to around 75 ppbv at 9.0 km, from where it increased substantially to more than 150 ppbv at 13.5 km above ground and higher values at the higher altitudes. Note that the sharp O_3 increase between 6.0 km and 13.5 km altitudes closely followed the increase of potential temperature.

At southwestern China over Kunming, O_3 mixing ratio showed a steady increase from the surface to around 13.5 km from where it started to increase sharply. The increase from surface to 13.5 km is slow. The sharp increase from 13.5 km coincided with an increase in the potential temperature, which indicates the tropopause. The O_3 mixing ratio below 13.5 km ranged from 50 to 75 ppbv. We noted that there was a distinguishable local O_3 peak with higher O_3 at the 3.0-4.5 km altitude. At southeastern China over Hong Kong, the O_3 profile showed a more complex manner. It showed a fairly sharp increase from the surface to reach a distinct local peak at around 3.0 km with a mixing ratio of 74 ppbv. The O_3 mixing ratio remained at around 60-70 ppbv up to 10.5 km from where it showed a steady decreasing trend to reach a minimum of around 50 ppbv at around 14.0 km. It then increased sharply at higher altitudes.

We have compared the averaged O_3 profile observed over Hong Kong in 2001 to the monthly average profiles observed from October 1993 to December 2001. We noted that the low O_3 mixing ratio at the upper troposphere over Hong Kong in March and April 2001 was not usually observed in the same months in other years. The O_3 mixing ratio in March and April usually shows a steady increasing trend from the middle to upper troposphere (Figure 3). Instead the general appearance of the averaged profile in 2001 looked like those observed in the wintertime, February and December for instance. One exception is the similarity between the local O_3 peaks over the lower troposphere of Hong Kong and Kunming which looked quite like that of the averaged enhancement O_3 profile reported by *Chan et al.* [2002] at the lower troposphere (2.0-6.0 km) from late February to May between 1994 and 1999 over Hong Kong (Figure 4).

Source and Origin of Ozone in the Upper Troposphere

Chan et al. [1998] attributed the occurrence of low O_3 in late autumn and wintertime over the upper troposphere over Hong Kong to the injection of O_3-depleted air masses from the tropical low-level convergence zone near the Indonesian region following the convective arm of the East Asian local Hadley circulation. This kind of transport of O_3-depleted air mass has been confirmed by the O_3 and trace gas measurements on board the DC-8 aircraft during the PEM-West B experiment [*Newell et al.*, 1997, *Kawakami et al.*, 1997]. Figure 5 shows the streamlines and isotachs at 150 (~ 14 km), 200 (~ 12 km) and 300 (~ 9.5 km) hPa altitudes averaged over March 1 to April 13, 2001. Westerly wind dominated most of Asia and western Pacific during the TRACE-P period. However, at higher altitudes especially the 150 and 200 hPa (12 km) there was an obvious anticyclonic flow bringing tropical air from the equatorial Pacific near the inter-tropical convergence zone. The flow influenced most Indo-China regions of SE Asia, the South China Sea and part of subtropical southeastern China including Hong Kong and Taiwan. Tropical air is rich in moisture content. In fact, the transport of the moisture-rich air had been reflected by a sudden jump in the water mixing ratio profile over Hong Kong around 9.0 km (Figure 2). Judging from the profiles at Kunming and LinAn, such kind of transport process did not affect the tropospheric O_3 over southwestern China and edge of subtropics at central-eastern China. This is consistent with the flow pattern in Figure 5 that the influence of the tropical air became diminished at higher latitudes, where it was dominated by a westerly flow.

The simultaneous increases of potential temperature and O_3 in the upper troposphere over LinAn and the differences in the O_3 profile of this site from that of Hong Kong suggest that there is a different source contributing to the O_3 in the upper troposphere of LinAn. Simultaneous increases in O_3 and potential temperature in the troposphere is often considered to be an indication of influences of stratospheric O_3 in the troposphere [*Appenzeller and Davies*, 1992]. LinAn is located at the edge of the subtropics near the middle latitude East Asian coast, where the strongest jet stream among the globe is found in the middle and upper troposphere in springtime [*Austin and Midgley*, 1994]. This jet stream is a steady feature of the winter circulation, which intensifies over eastern China and Japan and weakens in the western Pacific. The entrance region of the jet is to the east of 140^{o}E and the exit region is to the west of 140^{o}E [*Ding*, 1994]. The jet core of this stream in the TRACE-P period stretched west from over South Japan to over central-eastern China coast including LinAn (Figure 5). The wind speed of the jet stream decreased rapidly south of the jet core over Hong Kong and Kunming. The influence of this jet stream over LinAn is reflected by the maximum wind speed (~ 75 m/s) at around 12.0 km altitude (Figure 2). The average maximum wind speed over Hong Kong is around 30 m/s.

Previous studies in Japan have suggested that the jet stream very often is associated with transport of stratospheric O_3 into the troposphere through tropopause folding and subsequent diffusion processes [*Muramatsu et al*., 1984, *Wakamatsu et al.*, 1989, *Austin and Midgley*, 1994, *Tsutsumi and Makino*, 1995]. *Austin and Midgley* (1994) noted that the stratospheric-tropospheric exchange of ozone over northern Japan in spring is associated with the advection of laminae of high and low ozone mixing ratio in the stratosphere. We have observed frequently a similar advection of laminae of high ozone mixing ratio from the lower stratosphere to the upper and middle troposphere over LinAn. Such a case is shown by the O_3 profiles in Figure 6 from March 10 to 13. Within this period, layers of air with high O_3 mixing (> 300 ppbv) advected progressively from the lower stratosphere above 16.0 km to around 15.0 km on March 10, 14.8 km (175 ppbv) on March 11 (175 ppbv) and 12 (125 ppbv). Note that there were elevated-O_3 layers at the middle and upper tropopshere between 8.0 and 15.0 km from March 10 and 13. These layers were probably the remains of similar advection of O_3 laminae in earlier periods.

We have examined the possible relationship between O_3 and potential vorticity (PV) over the middle and upper troposphere of LinAn during the experimental period. Figure 7 shows the vertical distribution of O_3 mixing ratio and PV calculated from NCEP reanalysis data for the whole experiment period. Potential vorticity is a dynamic tracer of stratospheric air in the troposphere [*Danielsen*, 1968]. Rather high mixing ratios were observed for O_3 and there is a prominent vertical variation from the lower stratosphere to the middle troposphere around 8 km ASL. Noticeably tongues of extremely high ozone (with a maximum O_3 mixing ratio up to 1200 ppbv) extended from the lower stratosphere deep into the upper and middle troposphere during the beginning, middle and end of the experiment period. Also, high PV with value greater than $10^{-7}s^{-1}$ m^2 kg^{-1} (1 PVU = $10^{-7}s^{-1}$ m^2 kg^{-1}) was found at altitudes higher than 8.0 km with an exception from March 6 to 8 and March 15 to 20. The extension of these tongues of high O_3 mixing ratio into the middle and upper troposphere always corresponded well with the PV. For instance, at altitudes from 12.0 to 14.0 km from March 20 to April 7, PV values greater than 5 were found at the centre of the peak ozone mixing ratio regions. The high PV values in the middle and upper troposphere over LinAn especially during the extreme high ozone periods highly suggested that the ozone is of stratospheric origin. The above findings lead us to believe that the high O_3 found in the upper troposphere over LinAn in spring 2001 was mainly from stratospheric origin. Since Hong Kong and Kunming are relatively far away from the jet stream, the contributions of stratospheric O_3 to the tropospheric O_3 were relatively small. This is supported by the poor relationship between O_3 and PV (not shown).

Biomass Burning in SE Asia as A Source of Ozone in the Lower Troposphere

The occurrence of a similar local O_3 peak at the lower troposphere over Hong Kong and Kunming suggests that there may be a common source leading to such O_3 maximum. Examination on the individual O_3 profile at the three launching sites in year 2001 suggests that elevated O_3 concentrations at the lower troposphere were frequently observed over Hong Kong, less often over Kunming and scarcely over LinAn. These O_3 profiles are very similar to those reported by *Liu et al.* [1998], *Chan et al.* [2001] and *Chan et al.* [2002]. *Chan et al.* [2001] and *Chan et al.* [2002] showed that the O_3 enhancement are due to the transport of photochemical O_3 produced from the biomass burning emissions from the SE Asian subcontinent.

Chan et al. [2002] used the fire counts deduced from Along Track Scanning Radiometer (ASTR) satellite imagery from the European Space Agencyto show that biomass burning activities in the SE Asian subcontinent reached their full strength in the February-April period. In year 2001, active biomass burning activities also occurred in the SE Asian subcontinent. *Heald et al.* [this issue] estimated using Advanced Very High Resolution Radiometer data that burning in February-April 2001 was 87% of the climatological average reported by *Duncan et al.* [2002]. These active fire activities were commonly found in the Indo-Burma region containing Burma and Laos and Indo-China Indo-China region containing northern Thailand. Images from the ATSR World Fire Atlas (http://shark1.esrin.esa.it/ionia/FIRE/AF/ATSR) (Figure 8) showed that such fire activities resulted in the emission of O_3 precursors such as carbon monoxide (CO), methane, nonmethane hydrocarbons and others into the atmosphere of nearby regions. In fact, elevated CO column had been detected by the MOPITT (Measurements Of Pollution In The Troposphere) satellite (http://www.eos.ucar.edu/mopitt/dataimages/index.html) over the Indo-Burma region of SE Asia close to the Yunan Province, where the Kunming launching site is situated (Figure 9).

The emissions from the burning activities are believed to cause enhancements on tropospheric O_3 in the downwind subtropical Chinese mainland and the western Pacific. An obvious enhancement case observed from February 21 to March 13, 2001. Figure 10 shows the O_3 profiles over Hong Kong, Kunming and LinAn within the period. An elevated O_3 layer with peak concentration at around 75 ppbv started to occur on February 21 and 27 at 2.0 - 3.8 km altitude over Hong Kong. The peak concentration gained in strength on March 2 and reached the full strength with O_3 mixing ratio around 105 ppbv on March 7 before losing strength on March 9. Over Kunming a similar enhanced O_3 layer was not observed until March 8, when O_3

reached a maximum of 100 ppbv at 4.4 km altitude. The enhanced O_3 layer however lasted only for a short period of time till March 10, when much lower O_3 values were found. The O_3 profiles over LinAn showed no noticeable enhancement layer at similar altitudes.

Figure 11 shows the geopotential heights and streamlines for March 7 and 8. It indicates that there were subtropical anticyclones over the northern Philippines and the South China Sea on March 7 and 8, 2001, which resulted in an anticyclonic flow within the region from 5-30^{o}N to 90-140^{o}E. Such flow should have enabled transport of the biomass burning emissions from the SE Asian subcontinent to the downwind Pacific including the nearby vicinity of the SE Asia portion in China including Kunming and Hong Kong. The back air trajectories ending at the O_3 peak altitudes over Hong Kong and Kunming (not shown) tracked back to the Indo-Burma and Indo-China regions, where elevated CO column was observed on March 4 by the MOPITT (Figure 9), several days ago before the air masses reaching these sites. We thus believe that the O_3-enhanced layers over the lower troposphere of Hong Kong and Kunming are similarly caused by the biomass burning emissions from the SE Asian subcontinent, as were shown in earlier study for Hong Kong [*Chan et al.*, 2001, *Chan et al.*, 2002]. The trajectory at similar altitudes over LinAn (not shown) however, showed that the air masses were mainly from the western part of Chinese mainland. They are believed to be relatively free from the influence of the SE Asian biomass burning emissions.

CONCLUSION

During the TRACE-P period in spring 2001, we launched ozonesonde at three subtropical Chinese sites at Kunming of southwestern China close to the SE Asian border, Hong Kong in the southeastern Asia coast, and LinAn at the edge of subtropics of central-eastern China. The results showed that there are substantial variations in the vertical distributions over these subtropical sites. There were usually low O_3 values at the upper troposphere of Hong Kong, high O_3 values at the middle and upper troposphere of LinAn and frequent O_3 enhancements at the lower troposphere of Hong Kong and Kunming. The results were analysed by comparing these ozonesonde data to the long-term data from Hong Kong. We also used PV and metrological analysis, and fire observations and column carbon monoxide information from satellite observations to trace the origins and sources of O_3. The low values over the upper troposphere of Hong Kong is the result of the transport of O_3-depleted air from the inter-tropical convergence zone of equatorial regions following the East Asia local Hadley circulation. Such transport process does not affect the higher latitude at the edge of subtropics of LinAn where stratospheric O_3 is the major contributing source to the middle and upper tropospheric $O_{3.}$ The O_3 enhancements over the lower troposphere of Kunming and Hong Kong are caused by the SE Asian biomass burning emissions. Such enhancements are frequently observed over Hong Kong, less often over Kunming and scarcely over LinAn, which means that the biomass burning emissions from SE Asia in spring 2001 had mainly affected the southern parts of subtropical Chinese region.

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FIGURE CAPTION

Figure 1. Map showing the ozoensodne launching sites in Kunming, Hong Kong and LinAn.

Figure 2. The average profile of ozone and water mixing ratio, potential temperature, wind speed and direction over Kunming (February 28 to April 13), Hong Kong (March 2 to April 25) and LinAn (February 21 to April 13).

Figure 3. The monthly average ozone profile for December (purple), February (red), March (blue) and April (dark) over Hong Kong from October 1993 to December 2001.

Figure 4. The average ozone (solid line), temperature (dash-dot line) and relative humidity (dot line) profiles for ozone enhancement cases for the period October 1993 to September 1999.

Figure 5. The average streamlines and isotachs (m/s) at 300, 200 and 150 hPa from March 1 to April 13, 2001.

Figure 6. The profile of ozone over LinAn from March 10 to 13, 2001.

Figure 7. Time-height contour plot of ozone mixing ratios (ppbv) potential vorticity (PVU, 1 PVU = 10 $*10^{-7}s^{-1} m^2 kg^{-1}$) between 6 and 18 km ASL over LinAn from March 3 to April 13.

Figure 8. Geographical distributions of fires in Southeast Asia in March and April 2001. The fire locations at a resolution of 1×1 km^2 are denoted in red.

Figure 9. The total column carbon monoxide on March 4, 2001.

Figure 10. The vertical profiles of ozone over (a) Hong Kong, (b) Kunming and (c) LinAn from February 21 to March 13, 2001.

Figure 11. The average streamlines and isotachs (m/s) at 700 hPa on March 7 and 8, 2001.